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# Impact of the assimilation of ozone from the Tropospheric Emission Spectrometer on surface ozone across North America

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[1] We examine the impact of assimilating ozone observations from the Tropospheric Emission Spectrometer (TES) on North American surface ozone abundances in the GEOS-Chem model in August 2006. The assimilation reduces the negative bias in the modeled free tropospheric ozone, which enhances the ozone flux into the boundary layer. Surface ozone abundances increased by as much as 9 ppb in western North America and by less than 2 ppb in the southeast, resulting in a total background source of ozone of 20–40 ppb. The enhanced ozone in the model reduced the model bias with respect to surface ozone observations in the western USA, but exacerbated it in the east. This increase in the bias in the boundary layer in the east, despite the agreement between the assimilation and ozonesonde measurements in the free troposphere, suggests errors in the ozone sources or sinks or in boundary layer mixing in the model. **Citation:** Parrington, M., D. B. A. Jones, K. W. Bowman, A. M. Thompson, D. W. Tarasick, J. Merrill, S. J. Oltmans, T. Leblanc, J. C. Witte, and D. B. Millet (2009), Impact of the assimilation of ozone from the Tropospheric Emission Spectrometer on surface ozone across North America, *Geophys. Res. Lett.*, 36, L04802, doi:10.1029/2008GL036935.

## 1. Introduction

[2] Tropospheric ozone ( $O_3$ ) is an important trace gas which significantly impacts air quality and climate. It is produced by the oxidation of carbon monoxide (CO) and VOCs in the presence of nitrogen oxides ( $NO_x$ ), with transport from the stratosphere providing an additional source. Precise estimates of the tropospheric  $O_3$  budget are desirable in an air quality context as  $O_3$  produced in

the free troposphere and the intercontinental transport of  $O_3$  provides a background contribution to  $O_3$  abundances at the surface. It has been estimated [e.g., Fiore *et al.*, 2002; Vingarzan, 2004] that this background source contributes as much as 20–45 ppbv to surface  $O_3$ . Fiore *et al.* [2002] showed that Asian and European anthropogenic emissions increase surface  $O_3$  in North America by 4–7 ppbv. Jacob *et al.* [1999] and Lin *et al.* [2008] have suggested that future increases in Asian anthropogenic emissions could provide an additional increase of 1–7 ppbv in surface  $O_3$  in North America. Accurately quantifying the contribution of background  $O_3$  to regional air quality is clearly critical for assessing air quality regulation strategies. However, a major challenge is the significant spatial and temporal variability of the sources and sinks of  $O_3$  in the free troposphere.

[3] Recently, Parrington *et al.* [2008] showed that assimilation of tropospheric  $O_3$  data from the Tropospheric Emission Spectrometer (TES) instrument into a chemical transport model (CTM) can significantly improve the simulation of  $O_3$  in the free troposphere. They showed that assimilation of TES data reduced from –35% to less than –5% the mean bias between free tropospheric  $O_3$  in the GEOS-Chem model and ozonesonde measurements from the IONS-06 (INTEX Ozonesonde Network Study) experiment [Thompson *et al.*, 2008]. We show here that the improved simulation of  $O_3$  in the free troposphere following assimilation of the TES data enhances North American surface  $O_3$  abundances through changes in the flux of  $O_3$  into the planetary boundary layer (PBL).

## 2. Methodology

### 2.1. TES Ozone Data

[4] The TES instrument [Beer *et al.*, 2001] is a high-resolution imaging infrared Fourier-transform spectrometer, launched aboard the NASA EOS Aura satellite in July 2004. The Aura satellite is in a polar Sun-synchronous orbit with a repeat cycle of 16 days. The instrument employs a nadir-viewing geometry with an instrument field-of-view at the surface of 8 km × 5 km. The data used in this study were measured using the global survey mode, in which the observations are taken every other day with a spacing of about 220 km along the orbit track. Trace gas profiles are retrieved using an optimal estimation approach as described by Bowman *et al.* [2006]. We use version V002 of the data, which are filtered using the mean and root mean square of the radiance residual and on the cloud top pressure of each profile as recommended in the TES L2 Data User's Guide [TES Science Team, 2006]. These data have been validated by Nassar *et al.* [2008] who estimate that the TES  $O_3$  retrievals are biased high, in comparison to ozonesondes, by

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between 2.9 and 10.6 ppbv in the upper troposphere, and 3.7 and 9.2 ppbv in the lower troposphere. As given by Parrington *et al.* [2008], the TES retrievals used in this analysis have peak sensitivity to tropospheric O<sub>3</sub> at 700 hPa and 400 hPa, which provides valuable constraints on free tropospheric O<sub>3</sub>.

## 2.2. GEOS-Chem Model

[5] GEOS-Chem [Bey *et al.*, 2001] is a global 3-D CTM driven by assimilated meteorological data from the NASA Goddard Earth Observing System (GEOS-4) from the Global Modeling and data Assimilation Office (GMAO). The model includes a detailed description of tropospheric O<sub>3</sub>-NO<sub>x</sub>-hydrocarbon chemistry. The model has been used in a range of studies, including the analysis of the impact of background O<sub>3</sub> on surface O<sub>3</sub> over North America [e.g., Fiore *et al.*, 2002] and interpretation of in situ aircraft observations from the International Consortium on Atmospheric Transport and Transformation (ICARTT) aircraft campaign over North America [e.g., Hudman *et al.*, 2007]. We use version v7-02-04 of GEOS-Chem with a horizontal resolution of 2° × 2.5° and 55 levels in the vertical from the surface to 0.01 hPa, with 5 levels in the PBL. The emission inventories employed in the model are as described by Parrington *et al.* [2008]. However, we have adopted here the recommended updates to the US Environmental Protection Agency (EPA) National Emission Inventory 1999 (NEI99) from Hudman *et al.* [2008]. NO<sub>x</sub> emissions from industry and power plants have been reduced by 50% and anthropogenic emissions of CO have been reduced by 60%, relative to the NEI99 values.

## 2.3. Data Assimilation Framework

[6] Profiles of ozone and carbon monoxide from TES are assimilated into GEOS-Chem using a sequential Kalman filter as described by Parrington *et al.* [2008]. The TES profile retrievals are ingested into the model along the orbit track within each assimilation window of 6 hours to compute an analysis profile  $\hat{\mathbf{x}}^a$ :

$$\hat{\mathbf{x}}^a = \mathbf{x}^f + \mathbf{K}(\hat{\mathbf{x}}^{obs} - \mathbf{H}\mathbf{x}^f) \quad (1)$$

where  $\mathbf{H}$  is the observation operator,  $\mathbf{x}^f$  is the model (or forecast) profile,  $\hat{\mathbf{x}}^{obs}$  is the retrieved TES profile, and  $\mathbf{K}$  is the Kalman gain matrix, which is defined as:

$$\mathbf{K} = \mathbf{P}^f \mathbf{H}^T (\mathbf{H} \mathbf{P}^f \mathbf{H}^T + \mathbf{R})^{-1} \quad (2)$$

where  $\mathbf{P}^f$  is the error covariance matrix of the forecast profile and  $\mathbf{R}$  is the observation error covariance matrix provided with the TES retrievals. The assimilation was performed between 1 July through to 31 August 2006 with an assumed initial forecast error of 50% of the initial forecast field which we assume also captures the representativeness error. It is important to note, as reported by Parrington *et al.* [2008], that the assimilation approach employed here is suboptimal in that it neglects horizontal correlations in the forecast error covariance matrix, which would help spread the information from the TES retrievals. Since we do not account for the high bias in the TES retrievals, the agreement between the assimilation and the ozonesondes reported by Parrington *et al.* [2008] suggests

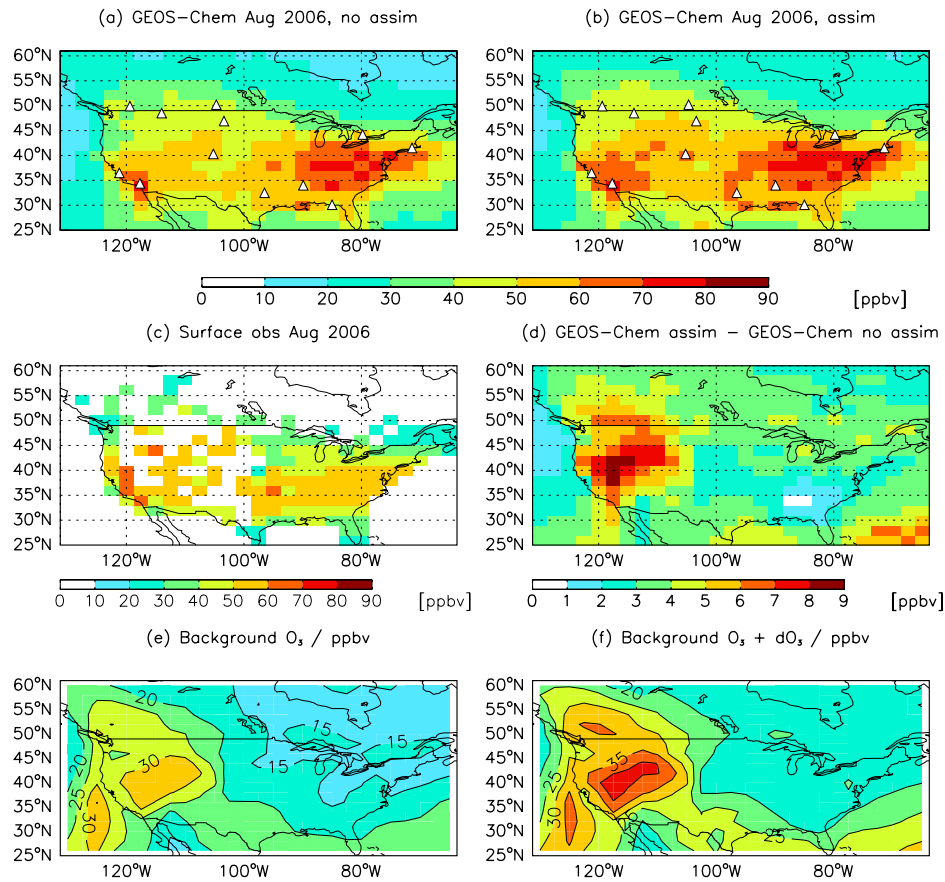
that the impact of the bias in the TES data is masked by the neglect of the horizontal correlations in the assimilation. Vertical correlations due to the smoothing influence of the TES retrievals are accounted for in the forecast error covariance matrix through the influence of the averaging kernels in the observation operator  $\mathbf{H}$ .

## 3. Results

[7] The results presented here are based on the assimilation results of Parrington *et al.* [2008], in which TES data for July and August 2006 were assimilated into the GEOS-Chem model. Figure 1 shows monthly afternoon (12 h to 18 h local time) averages of surface O<sub>3</sub> for August 2006. Comparison of the modeled surface O<sub>3</sub> distribution before assimilation (our standard simulation, shown in Figure 1a) with surface measurements (Figure 1c) (hourly data from the EPA Air Quality System (AQS) [U.S. Environmental Protection Agency, 2008] and the Environment Canada (EC) National Air Pollution Surveillance (NAPS) (<http://www.etc-cte.ec.gc.ca/NapsAnnualRawData/Default.aspx>) networks) reveals that the model overestimates O<sub>3</sub> in the east and underestimates it in the west. The overestimate of O<sub>3</sub> in the east is discussed by Fiore *et al.* [2002] and is potentially due to a combination of errors in precursor emissions or in the PBL heights being too low, or due to the coarse model resolution leading to excessive NO<sub>x</sub> dilution overestimating O<sub>3</sub>. Changes in surface O<sub>3</sub> following assimilation, shown in Figure 1d, are small (increases of less than 2 ppbv) in the southeastern USA and large (increases of up to 9 ppbv) in western North America.

[8] To assess the direct impact of the assimilation on O<sub>3</sub> in the PBL in the model we conducted a sensitivity test in which the analysis increments below 800 hPa were set to zero so that only the modeled O<sub>3</sub> abundances in the free troposphere were adjusted. We found that the O<sub>3</sub> differences shown in Figure 1d changed by less than 1 ppbv, suggesting that the O<sub>3</sub> differences are indeed due to an increase in the flux of free tropospheric O<sub>3</sub> into the PBL rather than direct adjustment of O<sub>3</sub> in the PBL in the assimilation. Examination of the vertical flux of O<sub>3</sub> over western North America (between 30°–55°N and 100°–125°W), for example, showed that the mean downward flux of O<sub>3</sub> into the PBL in this region increased by 71% following assimilation from  $2.5 \times 10^{10}$  molec. cm<sup>-2</sup> s<sup>-1</sup> to  $4.2 \times 10^{10}$  molec. cm<sup>-2</sup> s<sup>-1</sup>.

[9] Figure 1e shows the contribution of background O<sub>3</sub> to surface O<sub>3</sub> over North America, estimated using a tagged O<sub>3</sub> simulation with GEOS-Chem. In the tagged O<sub>3</sub> simulation the O<sub>3</sub> chemistry is linearized using 24-hr averaged production rates and loss frequencies for odd oxygen (O<sub>x</sub>) archived from the standard model simulation. Total O<sub>x</sub> is defined as (O<sub>3</sub> + NO<sub>2</sub> + 2NO<sub>3</sub> + PAN + PMN + HNO<sub>4</sub> + 3N<sub>2</sub>O<sub>5</sub> + HNO<sub>3</sub>). The background O<sub>3</sub> is defined as O<sub>3</sub> produced outside the North American PBL (15°–70° N, 125°–65° W, and from the surface to 750 hPa) and is calculated using a separate tracer. The background O<sub>3</sub> values in the standard simulation estimated from the tagged O<sub>3</sub> tracer, shown in Figure 1e, range from 15 ppbv in the east to 25–30 ppbv in the west. Background O<sub>3</sub> abundances, and thus the changes in surface O<sub>3</sub> following assimilation, are largest in the west due to the fact that the depth of the PBL is generally higher [e.g., Fiore *et al.*, 2002; Dougherty, 2008] and the O<sub>3</sub>



**Figure 1.** Monthly mean surface  $O_3$  (12 h–18 h local time) over North America for August 2006. GEOS-Chem surface  $O_3$  from the (a) standard and (b) assimilated simulations respectively, with the (d) difference between the two. (c) Surface  $O_3$  measurements from the EPA AQS and Environment Canada NAPS measurement networks, averaged on the  $2^\circ$  latitude  $\times$   $2.5^\circ$  longitude GEOS-Chem grid. (e) Contribution of background  $O_3$  to North American surface  $O_3$  and (f) combined contribution of the background  $O_3$  and the change following the TES assimilation.

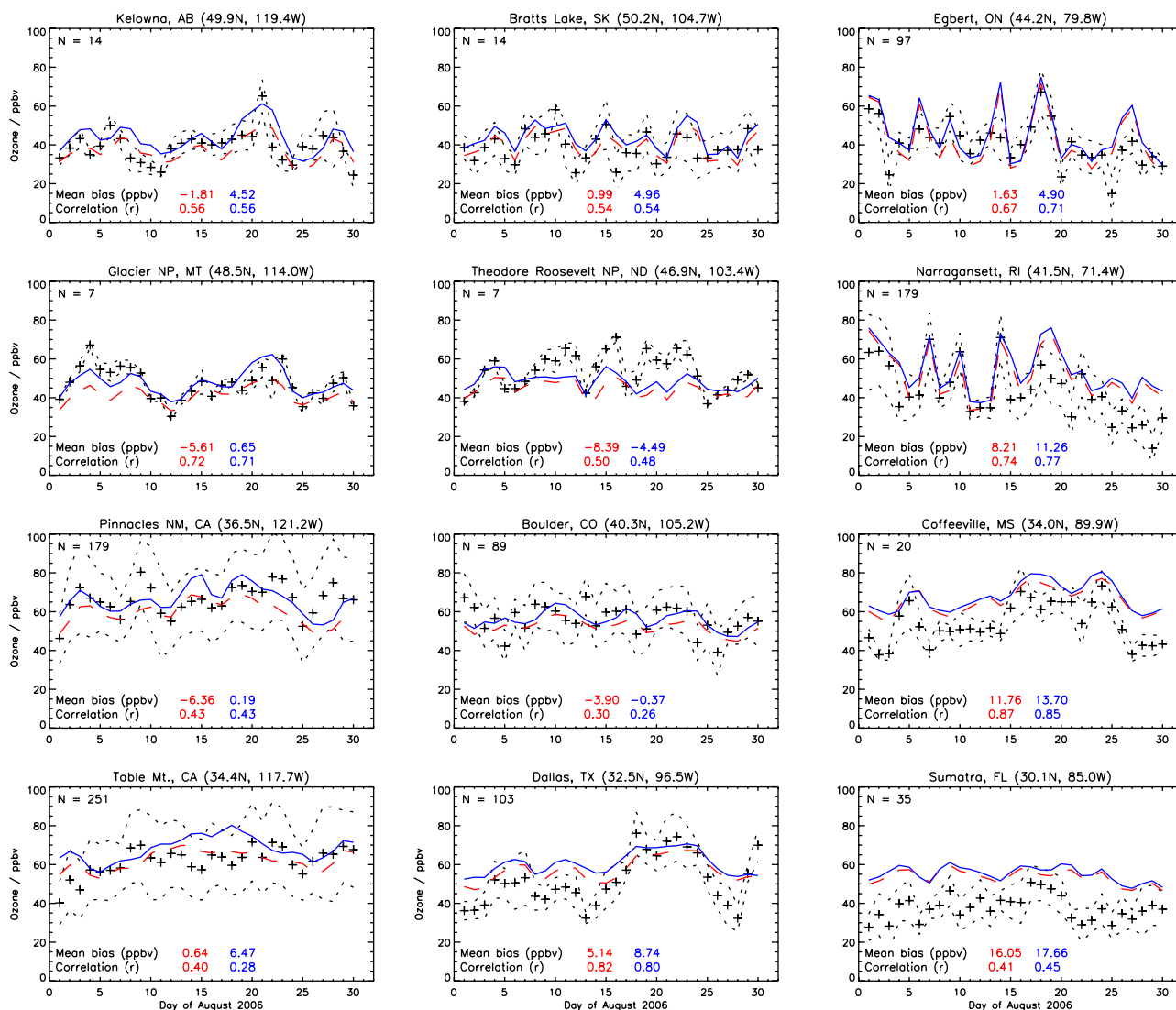
lifetime is longer [e.g., Fiore *et al.*, 2002] in the west than in the east. The total background  $O_3$  abundance following assimilation (i.e., the tagged  $O_3$  background plus the increase in surface  $O_3$  due to the assimilation) is shown in Figure 1f. Background  $O_3$  values with the assimilation are about 20–40 ppb across North American, an increase of 20–30% over background  $O_3$  in the standard simulation. The abundances of background  $O_3$  estimated here are higher than the 15–35 ppb estimated by Fiore *et al.* [2002] for summer 1995 by an amount comparable to the increase in  $O_3$  implied by the trend in background  $O_3$  of 0.19–0.51 ppbv year<sup>−1</sup> reported by Jaffe and Ray [2007]. However, because of the differences in the versions of the GEOS-Chem model employed here and by Fiore *et al.* [2002], as well as the limited one-month analysis presented here, the differences between our estimate of background  $O_3$  and that of Fiore *et al.* [2002] cannot be used to reliably infer a trend in background  $O_3$ .

[10] In Figure 2 we compare the modeled surface  $O_3$  to the timeseries of surface measurements at a number of observation sites across North America (indicated as white triangles in Figures 1a and 1b). In the western USA (e.g., Glacier National Park, Pinnacles National Monument, and Boulder) the assimilation improves the bias relative to the surface data. In western Canada (e.g., Kelowna and Bratt’s

Lake) and in the southwestern USA (e.g., Table Mt) the absolute mean bias between the model and observations is less than 2 ppbv, and is exacerbated following assimilation, resulting in an overestimate of surface  $O_3$  at these sites. At the eastern sites (e.g., Egbert and Narragansett), although GEOS-Chem overestimates surface  $O_3$  values and the assimilation increases the mean bias, there is good correlation ( $r > 0.7$ ) between the modeled and observed  $O_3$ . These results are consistent with the results of Tang *et al.* [2009] who found that using ozonesonde measurements as boundary conditions for a regional model significantly improved the modeled  $O_3$  in the free troposphere, but led to an overestimate of  $O_3$  in the PBL.

[11] Comparison of monthly mean model  $O_3$  profiles in the lower troposphere (below 500 hPa) with ozonesonde profiles from the IONS-06 campaign is shown in Figure 3. At all sites the model underestimates  $O_3$  in the free troposphere, but in the PBL the differences between the model and ozonesondes show the same geographical dependence as the timeseries plots, with  $O_3$  abundances underestimated in the west and overestimated in the east. In the free troposphere, the assimilation improves the  $O_3$  abundances at all sites. In the PBL, the assimilation reduces the mean bias in the model relative to the ozonesondes at the western sites, apart from Table Mt., where there is a





**Figure 2.** Time series of daily, afternoon (12 h–18 h local time), surface  $O_3$  at a number of surface measurement sites across North America. In each plot, the dashed red and solid blue lines show the standard model simulation and the assimilation respectively. Surface observations are shown by the black plus symbols, with the black dotted lines showing the one sigma standard deviation. The surface data represent all of the surface measurements within the gridbox coincident with the specified location, of which there are an average of  $N$  per day. The mean bias and correlation coefficient between the standard and assimilated model simulations, and the surface data are indicated in red and blue, respectively.

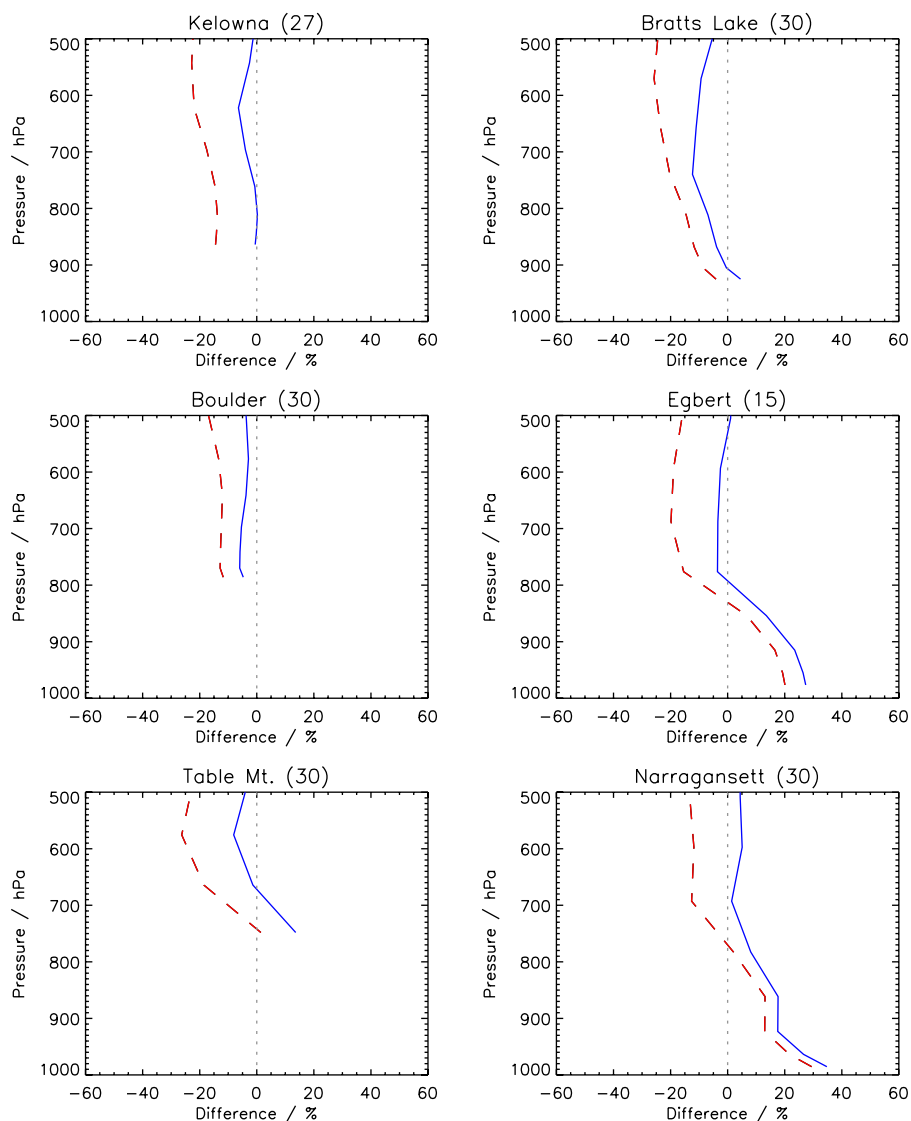
slight overestimate of  $O_3$  in the lowest levels of the model. At the eastern sites the  $O_3$  abundance is significantly improved above 800–700 hPa, but there is a slight increase in the bias in the PBL.

#### 4. Discussion

[12] We have presented an assessment of the response of summertime surface  $O_3$  abundances across North America in the GEOS-Chem model to the assimilation of tropospheric  $O_3$  observations from the TES satellite instrument. We showed that assimilation of TES data improves significantly the modeled abundances of  $O_3$  in the free troposphere relative to ozonesondes, resulting in an increase in the flux of background  $O_3$  into the North American PBL. Surface  $O_3$  in the model increased by 0–9 ppbv, with the largest increases in western North America, resulting in a total

contribution of background  $O_3$  to North American surface  $O_3$  of 20–40 ppb. Comparison of the modeled  $O_3$  abundances to observations of surface  $O_3$  from the EPA AQS and the EC NAPS networks showed that the model overestimates  $O_3$  in the PBL in eastern North America, but underestimates it in the west. Assimilation of TES data improved the modeled  $O_3$  at remote sites in the west, such as Glacier National Park and Pinnacles National Monument, but resulted in an overestimate of  $O_3$  at other western sites, such as Table Mt. and Kelowna. At all sites in the east, the small changes in surface  $O_3$  in the assimilation increased the model bias with respect to the surface measurements.

[13] The increase in the bias in surface  $O_3$  in the east and at some western sites following assimilation, despite the good agreement between the assimilated  $O_3$  and ozonesonde data in the free troposphere, indicates the presence of



**Figure 3.** Monthly mean percentage differences between GEOS-Chem, standard simulation (dashed red line) and assimilation (solid blue line), and ozonesonde profiles measured during the IONS-06 measurement campaign for August 2006. The number of ozonesonde profiles available for the comparison are given in brackets for each location.

model errors in the  $O_3$  sources or sinks, or in the simulation of the PBL mixing depths. This suggests that combining TES data with observations of  $O_3$  precursors, such as  $NO_2$  and  $HCHO$ , will enable us to more effectively isolate the impact on surface  $O_3$  abundances of discrepancies in local precursor emissions or in the description of transport into the PBL. It indicates that the TES data, when assimilated into a model of tropospheric chemistry and transport, can provide valuable constraints on the distribution of free tropospheric  $O_3$  as boundary conditions for the transport of background  $O_3$  into the PBL, which are critical when assessing the impact of air quality control strategies.

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ESRL Global Monitoring Division, and Environment Canada/MSC Ozone group.

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